Supplemental Material: High Temperature Superconductivity in $La_3Ni_2O_7$

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t_1^x	t_1^z	t_2^x	t_2^z	t_3^{xz}
-0.483	-0.110	0.069	-0.017	0.239
$t_\perp^x$	$t_\perp^z$	t_4^{xz}	ϵ^x	ϵ^z
0.005	-0.635	-0.034	0.776	0.409

TABLE I. Tight-binding hopping parameters in the main text Eq. (1) $H_(k)$ (unit here is eV). ϵ^x, ϵ^z are site energies for Ni$-d_{x^2-y^2}, d_{3z^2-r^2}$ orbitals, respectively.

sector	t_1^x	t_1^z	t_2^x	t_2^z	t_3^{xz}	ϵ^x	ϵ^z
ψ_S	-0.483	-0.110	0.069	-0.017	0.205	0.781	-0.226
ψ_A	-0.483	-0.110	0.069	-0.017	0.273	0.771	1.044

TABLE II. Tight-binding hopping parameters in ψ_S and ψ_A respectively.

Tight-binding parameters

The tight-binding hopping parameters for the bilayer two-orbital model in Eq. 1 of the main text are obtained from Ref.[1, 2]. Their parameters are listed in Tab.I.

Using the ψ_S and ψ_A orbitals, the $H(k)$ is block-diagonized into

$$H_{TB}(k) = \begin{pmatrix} H_S(k) & 0 \\ 0 & H_A(k) \end{pmatrix} \tag{1}$$

And $H_S(k)$, $H_A(k)$ take the same structure of $H_t(k)$

$$H_{S/A}(\mathrm{k}) = \begin{pmatrix} T_\mathrm{k}^x & V_\mathrm{k} \\ V_\mathrm{k} & T_\mathrm{k}^z \end{pmatrix}, \tag{2}$$

with $T_\mathrm{k}^{x/z} = t_1^{x/z}\gamma_k + t_2^{x/z}\alpha_k + \epsilon^{x/z}$, $V_\mathrm{k} = t_3^{xz}\beta_k$. Their parameters are listed in Tab.II.

For the fitting of the β band, we use up to the third nearest neighbor hopping in the TB model on the square lattice $t\gamma_k + t'\alpha_k + t''\gamma_{2k}$. The parameters are $t = 0.288$, $t' = -0.0746$, $t'' = 0.04$.

The coupled Hamiltonian for ψ_A and ψ_S

As described in the main text, the coupled Hamiltonian for ψ_A and ψ_S can be written as $H_\beta^{MF} + H_S^{MF} + H_{SAS}$, where H_β^{MF} is the mean-field Hamiltonian defined in Eq. (6) for the β band. The H_{SAS} is the coupling between two sectors in Eq. (7). The symmetric ψ_S sector is described by a two-band model with the exchange interaction,

$$H_S = \sum_{ij} t_{ij}^{\eta,\eta'} \psi_{\eta,i\sigma}^\dagger \psi_{\eta',j\sigma} + \sum_{\langle ij\rangle} J(\mathbf{S}_i \cdot \mathbf{S}_j - \frac{1}{4} n_i n_j) \tag{3}$$

Here $t_{ij}^{\eta,\eta'}$ are the hopping parameters in $H_S(k)$. The mean-field Hamiltonian H_S^{MF} follows from decoupling the exchange interaction into the two-orbital pairings and bonds as in Eq. (6). Notice that the band renormalization factors in $t_{ij}^{\eta,\eta'}$ are ignored because the ψ_S bands are heavily doped away from half-filling individually.

Hubbard Interactions in ψ_S and ψ_A

In this section, we discuss the interactions between ψ_S and ψ_A. Although the inversion symmetry blocks the hopping between ψ_S and ψ_A, the Coulomb interactions between them are nonzero and take a multiorbital form. More precisely, the local Hubbard interactions can be written as

$$\begin{aligned} H_I = {} & U \sum_{i,\eta} \hat{n}_{i,\eta\uparrow}\hat{n}_{i,\eta\downarrow} + U' \sum_{i,\eta\neq\eta'} \hat{n}_{i,\eta}\hat{n}_{i,\eta'} \\ & - J_H \sum_{i,\eta\neq\eta'} (\mathbf{S}_{i\eta} \cdot \mathbf{S}_{i\eta'} + d_{i,\eta\uparrow}^\dagger d_{i,\eta\downarrow}^\dagger d_{i,\eta'\uparrow} d_{i,\eta'\downarrow}) \end{aligned} \tag{4}$$

where the η is the orbital index.

Since the β band mainly carries the $d_{x^2-y^2}$ character, we will simply use the $d_{x^2-y^2}$ for ψ_β. Hence, the interaction between d_x^A and d_x^S coming from the intra-orbital $U\hat{n}_{i,\eta\uparrow}\hat{n}_{i,\eta\downarrow}$ takes the form

$$\begin{aligned} H_I^{A,S} = {} & U_0 \sum_{i,\alpha} \hat{n}_{i,\uparrow}^\alpha \hat{n}_{i,\downarrow}^\alpha + U_v \sum_{i,\alpha\neq\alpha'} \hat{n}_{i,\uparrow}^\alpha \hat{n}_{i,\downarrow}^{\alpha'} \\ & - J \sum_{i,\alpha\neq\alpha'} (d_{i,\uparrow}^{\alpha\dagger} d_{i,\downarrow}^{\alpha'\dagger} d_{i,\uparrow}^{\alpha'} d_{i,\downarrow}^{\alpha} + d_{i,\uparrow}^{\alpha\dagger} d_{i,\downarrow}^{\alpha\dagger} d_{i,\uparrow}^{\alpha'} d_{i,\downarrow}^{\alpha'}), \end{aligned} \tag{5}$$

where $\alpha = S, A$ and $U_0 = U_v = J = \frac{U}{2}$.

In the same spirit, we can decouple the inter-orbital interaction into a similar form. For example, the interaction between d_x^A and d_z^S comeing from $U'\hat{n}_{i,\eta}\hat{n}_{i,\eta'}$ takes the form

$$\begin{aligned} H_{I2}^{A,S} = {} & U_0 \sum_{i,\alpha} \hat{n}_{i,x}^\alpha \hat{n}_{i,z}^\alpha + U_v \sum_{i,\alpha\neq\alpha'} \hat{n}_{i,x}^\alpha \hat{n}_{i,z}^{\alpha'} \\ & - J \sum_{i,\alpha\neq\alpha'} (d_{i,x}^{\alpha\dagger} d_{i,z}^{\alpha'\dagger} d_{i,x}^{\alpha'} d_{i,z}^{\alpha} + d_{i,x}^{\alpha\dagger} d_{i,z}^{\alpha\dagger} d_{i,x}^{\alpha'} d_{i,z}^{\alpha'}) \end{aligned} \tag{6}$$

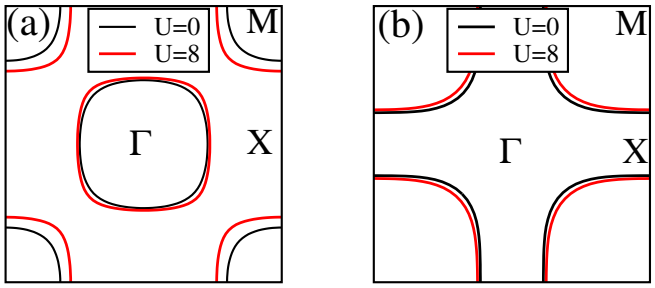

FIG. 1. (a) FSs of ψ_S at $U = 0$ (black lines) and $U = 8$ eV (red lines). (b) FSs of β band at $U = 0$ (black lines) and $U = 8$ eV (red lines).

with $\alpha = S, A$ and $U_0 = U_v = J = \frac{U'}{2}$. The Hund's rule interaction $J_H \mathbf{S}_{ix} \cdot \mathbf{S}_{iz}$ transforms into

$$H_{I3}^{A,S} = -J_0 \sum_i (\mathbf{S}_{ix}^S + \mathbf{S}_{ix}^A) \cdot (\mathbf{S}_{iz}^S + \mathbf{S}_{iz}^A) \quad (7)$$
$$-J_0 \sum_{i,\alpha\neq\alpha'} (d_{ix\sigma}^{A\dagger} d_{ix\sigma'}^S + d_{ix\sigma}^{S\dagger} d_{ix\sigma'}^A) \hat{\mathbf{S}}_{\sigma\sigma'} \cdot \hat{\mathbf{S}}_{\sigma'\sigma} (d_{iz\sigma'}^{A\dagger} d_{iz\sigma}^S + d_{iz\sigma'}^{S\dagger} d_{iz\sigma}^A)$$

with $J_0 = \frac{J_H}{2}$. Collecting all the terms, the symmetry allowed local interactions are just the multi-orbital Hubbard model with the effective orbitals including with both the atomic orbitals and the molecular symmetric-antisymmetric sector index. The inter-sector interactions are crucial and produce the inter-sector exchange interaction. As we discussed in previous works [3, 4], the inter-sector spin-orbital exchange interaction generates the effective Josephson coupling between the pairing order parameters,

$$H_{SAS} = J_{SA}(\hat{\Delta}_{Sx}^\dagger \hat{\Delta}_\beta + \hat{\Delta}_{Sz}^\dagger \hat{\Delta}_\beta + h.c.) \quad (8)$$

Finite-U Gutzwiller approximation

An important aspect of our theory is the doping concentration for the antisymmetric β band and the symmetric α and β bands. In the main text, we used the results of the DFT calculations, which are reproduced in the TB model. However, the strong local correlation can in principle generate inter-orbital and inter-sector charge transfer among the ψ_A and ψ_S bands by renormalizing the effective crystal fields. To this end, we carried out a finite-U multiorbital Gutzwiller approximation calculation [3, 4], including all four bands relevant for LNO. The results of the renormalized FSs are shown in Fig. 1 for the Hubbard interaction $U = 8$eV and Hund's coupling $J_H = 0.1U$ and compared to the noninteracting case. Clearly, the correlation-induced charge transfer is weak as indicated by the small changes in the sizes of the FSs for correlation strength up to $U = 8$eV, providing support for the results discussed in the main text.

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